TR-1753—A Two-Species Laser Model with Excitation Energy Transfer—by Nick Karayianis, Clyde A. Morrison, Donald E. Wortman

A Two-Species Laser Model with Excitation Energy Transfer

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A theoretical model is proposed for a laser system with two types of active ingredients--one with three energy levels that is capable of population inversion and a second that transfers energy to the former to aid in this inversion. Some properties of the model are examined under continuous-wave and Q-switched operations, to establish a theoretical insight regarding the behavior of multiply doped systems such as $LiYF_4$ doped with Ho, Tm, and Er. Concentration, temperature, and cavity Q dependences are introduced in

DD FORM 1 JAN 73 1473 EDITION OF 1 NOV 65 IS OBSOLETE

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered) simple, nontrivial ways, but are not fully explored in this preliminary report.

CONTENTS

FIGURES

1. INTRODUCTION

A promising solid-state laser material being investigated is $LiYF_4$ multiply doped with triply ionized holmium (Ho), thulium (Tm), and erbium (Er), the last two of which assist in selectively exciting the laser-active Ho. It is important to understand the role of Tm and Er from a quantitative point of view, to give some theoretical guidance to the optimum proportion of these dopants and to suggest other possible multiply doped systems.

This report is a preliminary theoretical incursion into this problem. A simple two-dopant model with phenomenological interactions is postulated. It displays the following properties that are observed in real systems:

(a) Population inversion between two levels of a given species A

(b) Excitation transfer from a second species B (donor) to the species A (acceptor)

(c) Concentration dependence of the behavior of the system

(d) Temperature effects

(e) Cavity-Q effects

Although in many instances the simplest nontrivial assumptions are made, the analysis is expected to develop some conceptual insights into the physical processes operative in an actual system and to stimulate a more detailed and physically related treatment. Accordingly, the best known functional dependence of some of the parameters on, say, concentration or temperature may not have been ascribed, but have been introduced phenomenologically to produce known results in limiting cases.

2. MODEL

Let us consider the two-species system shown in figure 1. The levels have been numbered sequentially for convenience of notation, although levels 1, 2, and 3 belong to species A, and levels 4 and 5 belong to species B. Let the time rate of change of the population of various levels be influenced by the following interactions:

(a) $-a_{ij}(n_i - n_j)$, stimulated emission and absorption between ith and jth levels,

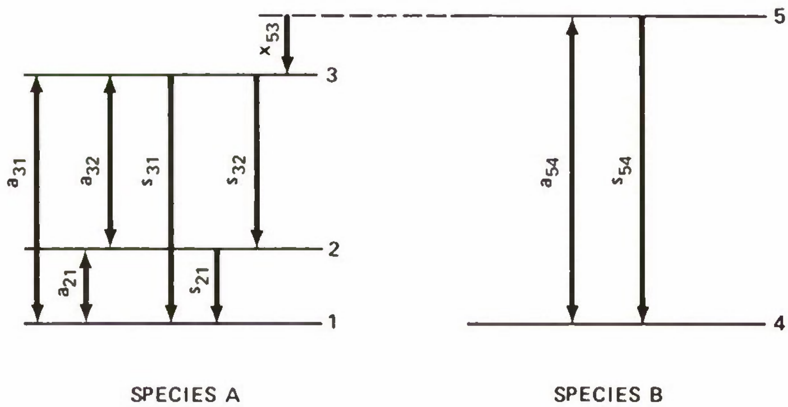

Figure 1. Population inversion between levels 3 and 2 is aided by excitation transfer between levels 5 (of species B) and 3 (of species A).

(b) $-s_{ij}n_i$, spontaneous emission from ith to jth level,

(c) $-x_{ij}n_i n_1$, excitation transfer from the ith level of species B to jth level of species A,

where

$$n_i = Nc_i f_i \tag{1}$$

is the number of occupied ith levels, N is the total number of sites capable of being occupied by either species A or B, c_i is the relative concentration of the species to which the ith level belongs, and f_i is the fractional population of the ith level. Thus,

$$c_1 = c_2 = c_3 = c_A \tag{2}$$

$$c_4 = c_5 = c_B \quad , \tag{3}$$

$$c_A + c_B \le 1 \quad , \tag{4}$$

$$f_1 + f_2 + f_3 = 1 \quad , \tag{5}$$

$$f_4 + f_5 = 1 \quad . \tag{6}$$

Conservation of species is assured by the rate equations, which are

$$\dot{n}_5 = -a_{54}\left(n_5 - n_4\right) - s_{54}n_5 - x_{53}n_5n_1 \quad , \tag{7a}$$

$$\dot{n}_4 = a_{54}\left(n_5 - n_4\right) + s_{54}n_5 + x_{53}n_5n_1 \quad , \tag{7b}$$

and

$$\dot{n}_3 = -a_{32}\left(n_3 - n_2\right) - a_{31}\left(n_3 - n_1\right) - \left(s_{32} + s_{31}\right)n_3 + x_{53}n_5n_1 \ , \quad (8a)$$

$$\dot{n}_2 = a_{32}\left(n_3 - n_2\right) - a_{21}\left(n_2 - n_1\right) - s_{21}n_2 + s_{32}n_3 \ , \quad (8b)$$

$$\dot{n}_1 = a_{31}\left(n_3 - n_1\right) + a_{21}\left(n_2 - n_1\right) + s_{21}n_2 + s_{31}n_3$$

$$- x_{53}n_5n_1 \ , \quad (8c)$$

whence

$$\dot{n}_5 + \dot{n}_4 = \dot{n}_3 + \dot{n}_2 + \dot{n}_1 = 0. \quad (9)$$

2.1 Stimulated Emission and Absorption Coefficient "a"

The stimulated emission and absorption term between levels i and j populates or depletes a level, depending on whether the population of the other level is greater or less than its own. When the lower energy level is more populated, there is a net absorption of any radiation of frequency ν_{ij}. The a_{ij} factors, among other things, are proportional to the radiation power available at the particular frequency ν_{ij}. For this model, levels 5 and 3 are assumed externally pumped, so that

$$a_{54} = P\alpha_{54} \ , \quad (10a)$$

$$a_{31} = P\alpha_{31} \ , \quad (10b)$$

where P is the external pump power.

As these two levels are pumped, some power appears in the system at the frequencies ν_{32} and ν_{21}. This power is assumed negligible, except for the power z of frequency ν_{32} in the laser mode to which the system is tuned with some Q. The time rate of change of the laser frequency power inside the cavity is

$$\dot{z} = N^{-1}b_{32}\left(n_3 - n_2\right)z + N^{-1}\eta s_{32}n_3 - \nu_{32}z/Q \quad (11)$$

As shown, the first term feeds or depletes the power, just as the stimulated emission and absorption term feeds the level populations. The second term represents the fraction η/N of the spontaneous emission at frequency ν_{32} from level 3 that goes into the laser mode. The third term represents the power lost per unit time and is inversely proportional to the cavity Q. Transmission loss of the laser mode within the cavity is assumed negligible, so that *all of the last term represents usable radiated power from the cavity.* Similar to equations (10a) and (10b),

$$a_{32} = z\alpha_{32} \ . \tag{12a}$$

For a_{21}, the following choice is made:

$$a_{21} = s_{21}\left[\exp\left(\nu_{21}/kT\right) - 1\right]^{-1} \tag{12b}$$

which maintains level 2 at the Boltzmann value when the pump power is turned off. All other levels are assumed sufficiently distant from their corresponding ground level to be negligibly populated at normal termperatures.

2.2 Spontaneous Emission Coefficient "s"

The spontaneous emission term between levels i and j depletes the upper level and populates the lower level. Spontaneous emission due to radiation and some nonradiative processes are implicitly contained in this term. All the s_{ij} are assumed to have a concentration dependence of the form

$$s_{ij} \approx \left[1 - \left(c_A + c_B\right)^M\right]^{-1} \tag{13}$$

where it is recalled by equation (4) that

$$c_A + c_B \leq 1.$$

This arbitrarily chosen dependence provides, phenomenologically, a quenching of the system due to coupling between ions as more activators B or lasing ions A are added. The optimum concentrations in this model obviously depend on the value chosen for M, but are clearly somewhat less than maximum.

In order that continuous-wave (CW) inversion can be established between levels 3 and 2 in this model, it is assumed that $s_{21} > s_{32} + s_{31}$. This assumption is consistent with the observed long lifetimes of fluorescing (lasing) levels.

2.3 Excitation Transfer Coefficient "x"

This model assumes excitation transfer occurs only from level 5 to level 3. The rate of transfer is assumed proportional to the population of level 5 and to the population of level 1. This excitation process implies that, simultaneously to the transition of an atom of species B from its excited state 5 to its ground state 4, a phonon of energy ν_{53} is created, and there is an excitation of an atom of species A from its ground state 1 to its excited state 3.

The neglect of reverse excitation in this model is tantamount to assuming that ν_{53}/kT is sufficiently large so the probability is essentially zero of reverse excitation of level 5 with the simultaneous annihilation of a phonon. In this model, some value is assigned to x where

$$x = Nx_{53}. \tag{14}$$

3. CONTINUOUS-WAVE OPERATION (STEADY-STATE SOLUTION)

Substituting $n_i = Nc_i f_i$ and $Nx_{53} = x$ into the linearly independent equations (7a), (8a), and (8b) gives for the steady-state equations

$$\dot{f}_5 = -a_{54}\left(f_5 - f_4\right) - s_{54}f_5 - xc_A f_5 f_1 = 0, \tag{15a}$$

$$\dot{f}_3 = -a_{32}\left(f_3 - f_2\right) - a_{31}\left(f_3 - f_1\right) - \left(s_{32} + s_{31}\right)f_3 + xc_B f_5 f_1 = 0 , \tag{15b}$$

$$\dot{f}_2 = a_{32}\left(f_3 - f_2\right) - a_{21}\left(f_2 - f_1\right) - s_{21}f_2 + s_{32}f_3 = 0, \tag{15c}$$

$$\dot{z} = b_{32}c_A\left(f_3 - f_2\right)z - \nu_{32}z/Q + \eta s_{32}c_A f_3 = 0 , \tag{15d}$$

which, together with

$$f_1 = 1 - f_2 - f_3 , \tag{16a}$$

$$f_4 = 1 - f_5 , \tag{16b}$$

$$a_{54} = P\alpha_{54} , \tag{16c}$$

$$a_{31} = P\alpha_{31} \ , \tag{16d}$$

$$a_{32} = z\alpha_{32} \ , \tag{16e}$$

solve the system.

The results are

$$f_5 = a_{54}\left[2a_{54} + s_{54} + xc_A f_1\right]^{-1}, \tag{17a}$$

$$\begin{aligned} f_1 = &\left[\left(a_{21} + s_{21}\right)(1 - u) - \left(s_{32} + \frac{G_1 s_{32} Q u}{1 - G_2 Q u}\right)(1 + u)\right] \\ &\times \left[3a_{21} + s_{21} - s_{32} - \frac{G_1 s_{32} Q u}{1 - G_2 Q u}\right]^{-1}, \end{aligned} \tag{17b}$$

$$z = \frac{G_1 s_{32} Q}{2\alpha_{32}} \left(\frac{1 + u - f_1}{1 - G_2 Q u}\right), \tag{17c}$$

$$f_2 = \left(1 - u - f_1\right)/2 \tag{17d}$$

$$f_3 = \left(1 + u - f_1\right)/2 \tag{17e}$$

$$G_1 = \eta\alpha_{32}/\nu_{32} \tag{17f}$$

$$G_2 = b_{32}c_A/\nu_{32} \tag{17g}$$

where the inversion, u, is defined by

$$u = f_3 - f_2 \ . \tag{18}$$

The inversion is related to P by

$$F(P,u) = 0 \tag{19a}$$

with

$$\begin{aligned} F(P,u) = &\left(2a_{54} + s_{54} + xc_A f_1\right)\left[a_{31}\left(1 + u - 3f_1\right) + 2\alpha_{32} z u \right. \\ &\left. + \left(s_{32} + s_{31}\right)\left(1 + u - f_1\right)\right] - 2xc_B a_{54} \ . \end{aligned} \tag{19b}$$

This expression (through a_{54} and a_{31}) is quadratic in P with the solution

$$P = -b_o + \left[b_o^2 + b_1\right]^{\frac{1}{2}} \quad , \tag{20}$$

where

$$\begin{aligned} b_o = & \left[4\alpha_{54}\alpha_{31}\left(3f_1 - 1 - u\right)\right]^{-1} \\ & \times\left\{2xc_B\alpha_{54}f_1 + \alpha_{31}\left(3f_1 - 1 - u\right)\left(s_{54} + xc_A f_1\right)\right. \\ & \left. - 2\alpha_{54}\left[2\alpha_{32}zu + \left(s_{32} + s_{31}\right)\left(1 + u - f_1\right)\right]\right\} , \end{aligned} \tag{21a}$$

$$\begin{aligned} b_1 = & \left[2\alpha_{54}\alpha_{31}\left(3f_1 - 1 - u\right)\right]^{-1} \\ & \times\left[s_{54} + xc_A f_1\right]\left[2\alpha_{32}zu + \left(s_{32} + s_{31}\right)\left(1 + u - f_1\right)\right]. \end{aligned} \tag{21b}$$

At $P = \infty$, $f_3 = f_1$, which implies that

$$3f_1 = 1 + u,$$

where f_1 is given in terms of u by equation (17b). The solution for u_{MAX}* is the smaller root of the quadratic

$$Au^2 - Bu + C = 0 \quad , \tag{22}$$

where

$$A = \left[G_2\left(3a_{21} + 2s_{11} + s_{32}\right) - G_1 s_{32}\right]Q \ , \tag{23a}$$

$$B = 3a_{21} + 2s_{21} + s_{32} + \left[G_2\left(s_{21} - s_{32}\right) + G_1 s_{32}\right]Q \quad , \tag{23b}$$

$$C = s_{21} - s_{32} \ . \tag{23c}$$

**u_{MAX}* means u evaluated at P_{MAX}, i.e., at $P = \infty$. It is later shown that maximum u is attained at $P = \infty$ only if $Q > Q_c$, where Q_c is defined in section 4.*

It has been assumed that $s_{21} > s_{31} + s_{32}$, which is a necessary condition for laser action, so each of these quantities is positive. Thus,

$$u_{MAX} = \frac{B}{2A} - \left[\left(\frac{B}{2A}\right)^2 - \frac{C}{A}\right]^{\frac{1}{2}} \tag{24}$$

gives a real positive solution, since it may be shown also that $B^2 > 4AC$. At $P = 0$, the population of level 3 vanishes ($f_3 = 0$), so from equation (17e),

$$f_1 = 1 + u\ ,$$

which gives

$$u_{MIN} = -\frac{a_{21}}{2a_{21} + s_{21}} = -\left[1 + \exp\left(\nu_{21}/kT\right)\right] \tag{25}$$

or from equation (12b),

$$f_2/f_1 = \exp\left(-\nu_{21}/kT\right) \tag{26}$$

as required.

As the pump power is increased from 0, u increases (not necessarily monotonically) from the negative value given by equation (25) to some Q-dependent positive value at $P = \infty$ given by equations (23) and (24). For $Q < Q_c$, u peaks at some power $P < \infty$ and then tends to u_{MAX} as follows:

$$u_{MIN} \leq u \rightarrow u_{MAX}(Q) \tag{27a}$$

for

$$0 \leq P \rightarrow \infty\ . \tag{27b}$$

The upper limit of u at $P = \infty$, $u_{MAX}(Q)$, varies with Q as

$$0 \leq u_{MAX} \leq \frac{s_{21} - s_{32}}{3a_{21} + 2s_{21} + s_{32}} \tag{28a}$$

for

$$\infty \geq Q \geq 0 \quad . \tag{28b}$$

Thus, this model displays accurately the known result that a greater population inversion can be obtained if the cavity Q is kept low during the pumping cycle. This property sets up the possibility of suddenly increasing Q, after a large inversion is set up to force the stored potential energy to appear as laser radiation at the resonant frequency. This procedure is appropriately referred to as Q-switching.

The exact point that may be designated as threshold is not too well defined. We define threshold as the point of inflection in the steep *rise* of z with pumping power. That is, $P = P_T$ is defined by

$$\frac{d^2z}{dP^2} = 0 \ ,$$

where

$$\frac{dz}{dP} > 0 \ . \tag{29}$$

Since z is a function of u and f_1, f_1 is a function of u, and u is related to P through equation (19), it is possible to express the second derivative of z in terms of the parameters of the system. However, the prospect is not too appealing of finding the appropriate zero of the resulting expression (even if the expression could be contained on one page). The procedure for solving the system is to assign values to u between the limits indicated by equation (27); calculate f_1 and then z from equations (17b) and (17c); determine P from equations (20), (21a), and (21b); and then evaluate f_5, f_2, and f_3 from equations (17a), (17d), and (17e), respectively.

4. CRITICAL CAVITY Q (Q_c)

Under CW operation, if Q is sufficiently large and the pump power is increased slowly, there is a value of $P = P_T$, the threshold power, for which a very rapid increase in the laser power, z, appears in the cavity. If, however, Q is less than some Q_c (derived below), there is no rapid buildup of z, no matter how large P becomes.

The peak value of u for $Q > Q_c$ is u_{MAX} given by equation (24), which obtains at $P = \infty$. For $Q < Q_c$, u peaks where the two roots of P are equal; that is, where

$$b_o^2 = -b_1 \tag{30}$$

in equation (20). For arbitrary Q, equation (29) is eighth degree in u and cannot be solved explicitly. However, $Q = Q_c$ implies that equation (29) is valid simultaneously to $u = u_{MAX}$ where $3f = 1 + u$. Letting this value of u be u_c, one obtains

$$u_c = \frac{s_{21} + s_{31} - xc_B/2}{3a_{21} + 2s_{21} - s_{31} + xc_B/2} \tag{31}$$

and

$$Q_c = u_c^{-1} \frac{s_{31} + s_{32} - xc_B/2}{G_2\left(s_{31} + s_{32} - xc_B/2\right) - G_1 s_{32}}, \tag{32}$$

where G_1 and G_2 are given by equations (17f) and (17g). For $Q < Q_c$, there is insufficient buildup of laser power to deplete the inversion, so u continues to increase with pump power to a value larger than $3f_1 - 1$, which implies $f_3 > f_1$. As the pump power is increased still further, it tends to deplete f_3 to establish $f_3 = f_1$ at $P = \infty$; thus, u decreases with power for $P > P_c$. This behavior is a consequence of the model, which assumes pumping directly into level 3, the lasing level.

For $Q < Q_c$, therefore, no laser action is possible in this system, whatever the pump power might be.

5. Q-SWITCHING (DYNAMICAL SOLUTION)

The behavior of this system under Q-switching is governed by the rate equations and the initial values of the parameters. To simplify notation, let

$$u = f_3 - f_2 \ , \tag{33a}$$

$$v = f_1 \ , \tag{33b}$$

$$w = f_5 \ , \tag{33c}$$

in which case equations (15) become

$$\begin{aligned}\dot{u} = & \left(a_{21} + s_{21} - a_{31} - s_{31} - 2s_{32}\right)/2 \\ & + \left(3a_{31} + s_{31} + 2s_{32} - 3a_{21} - s_{21}\right)v/2 \\ & + \left(a_{31} + s_{31} + 2s_{32} + a_{21} + s_{21}\right)u/2 \\ & + 2\alpha_{32}zu + xc_B wv \ ,\end{aligned} \tag{34a}$$

$$\begin{aligned}\dot{v} = & \left(a_{31} + s_{31} + a_{21} + s_{21}\right)/2 \\ & - \left(3a_{31} + s_{31} + 3a_{21} + s_{21}\right)v/2 \\ & + \left(a_{31} + s_{31} - a_{21} - s_{21}\right)u/2 \\ & - xc_B wv \ ,\end{aligned} \tag{34b}$$

$$\dot{w} = a_{54}(1 - 2w) - s_{54}w - xc_A wv \ , \tag{34c}$$

$$\dot{z} = b_{32}c_A zu - \nu_{32}z/Q + \eta s_{32}c_A\left(1 + u - v\right)/2 \ . \tag{34d}$$

These nonlinear, coupled equations are easily solved numerically by assigning values to u_o, v_o, w_o, and z_o at $t = 0$ and calculating the evolution of the system by $u_1 = u_o + \dot{u}\Delta t$, etc., for sufficiently small Δt.

Initial values may be obtained by setting $Q = 0$ and pumping at $P = P_o$, where P_o is the power for $Q = 0$ that gives the largest inversion. Then at $t = 0$, one may turn off the power and switch Q to some large value and see how the system evolves. It is found that if Q is kept constant after $t = 0$, the output power, $\nu_{32}z/Q$, peaks at later times, the larger the Q; and the peak power output is higher for some $Q > 0$, depending on the parameters of the system. If Q is switched back to $Q = 1$ when the power in the cavity peaks (it will peak at later times, the higher the Q), a large spike in output power occurs at that point. The *average* output power is greater, the greater the Q, but saturates at some $Q < \infty$.

6. RESULTS AND DISCUSSION

Figure 2 plots the inversion as a function of pump power for typical Q less than, equal to, and greater than Q_c. For Q-switching, maximum inversion is established by pumping at P_o with $Q = 0$. Figure 3 shows the variation of laser power z, in the cavity as a function of Q at infinite pump power. The "break" in the curve at $Q = Q_c$ indicates, as stated previously, that there is no appreciable buildup of laser power for Q's below this value.

This system was examined under two types (A and B) of Q-switching. In both types, maximum inversion was initially established by setting $P = P_o$ and $Q = 0$. In type A, the pump power was cut off, and Q was increased instantly to some positive value at $t = 0$ and kept constant thereafter. In type B, everything proceeded as in type A, except that when the cavity laser power reached its maximum, Q was lowered to $Q = 1$ (cavity dumping) and kept at this value thereafter. The time at which this lowering of Q (to dump the cavity power) occurs depends on the initial rise in Q--taking longer, the larger the Q.

Figure 4 shows the time variation of output power (the $\nu_{32}z/Q$ term in equation (11)) for Q-switching of type A. Peak output power occurs at later times for larger Q, and the largest pulse occurs at some intermediate Q value. Figure 5 shows the variation with Q of the peak output power and the time at which it obtains.

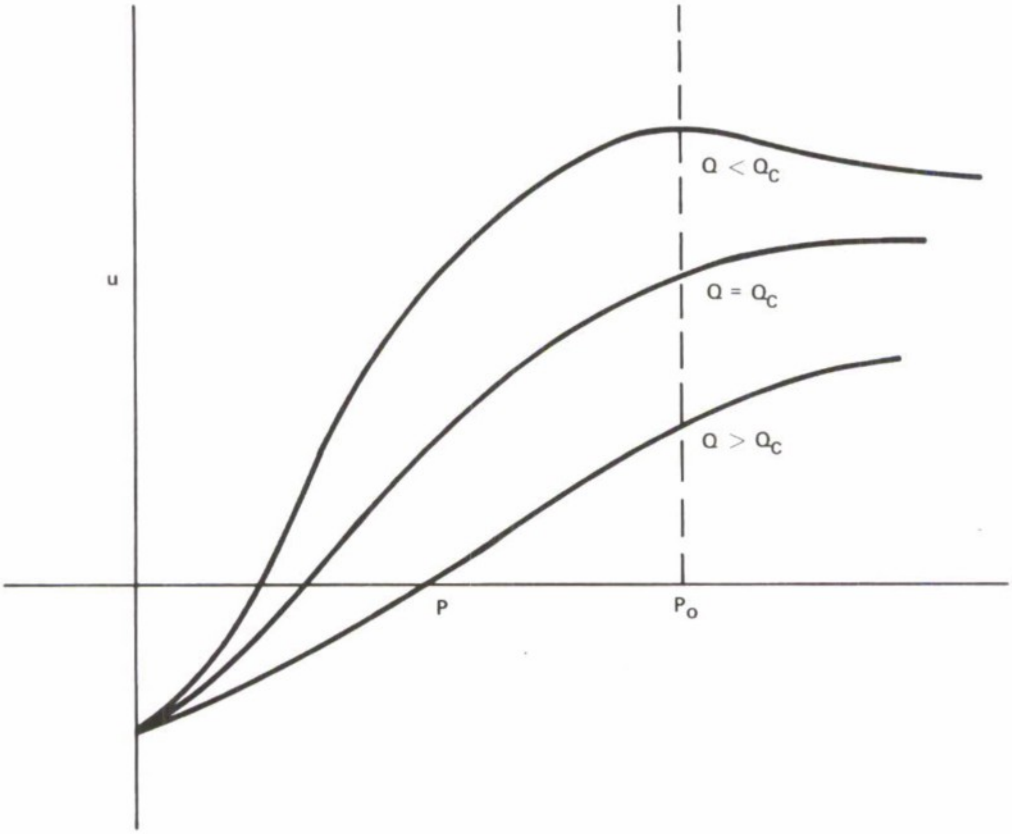

Figure 2. Inversion ($u = f_3 - f_2$) as a function of pump power P for Q <, =, > Q_c) the critical cavity Q, below which no laser action obtains.

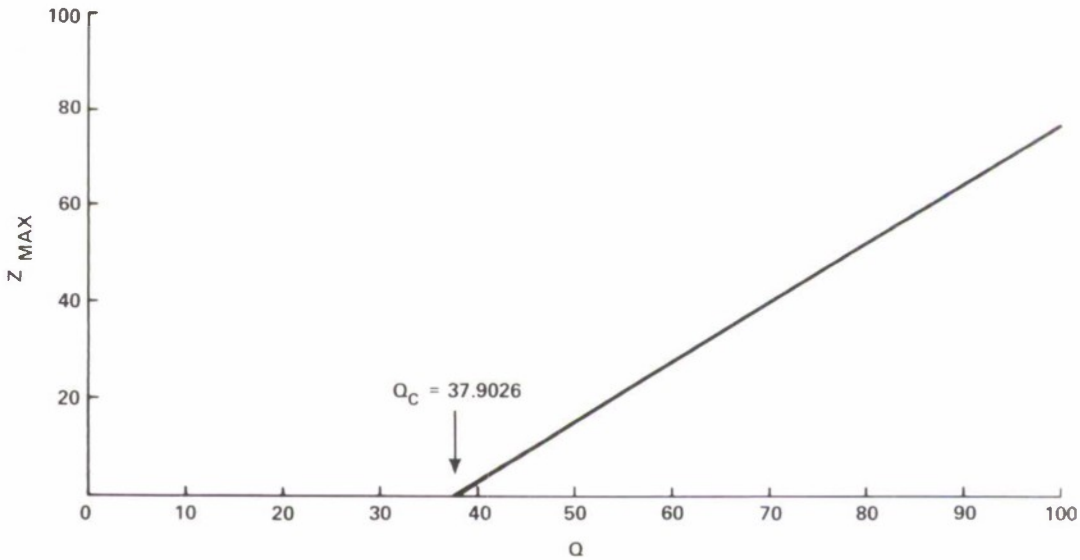

Figure 3. Laser power z in cavity versus cavity Q at infinite pump power (below $Q = Q_c = 37.9026$ there is no oscillation.

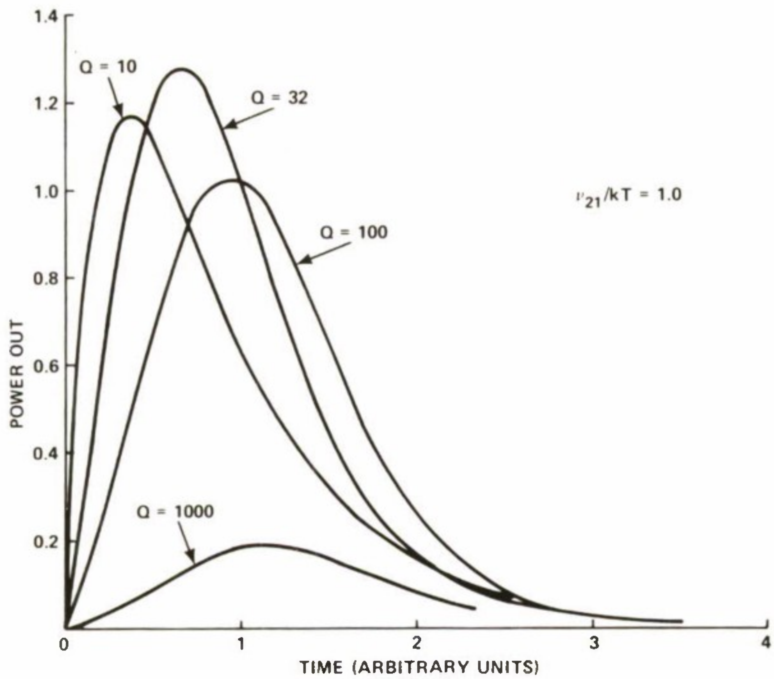

Figure 4. Power output versus time for several Q in type A Q-switching.

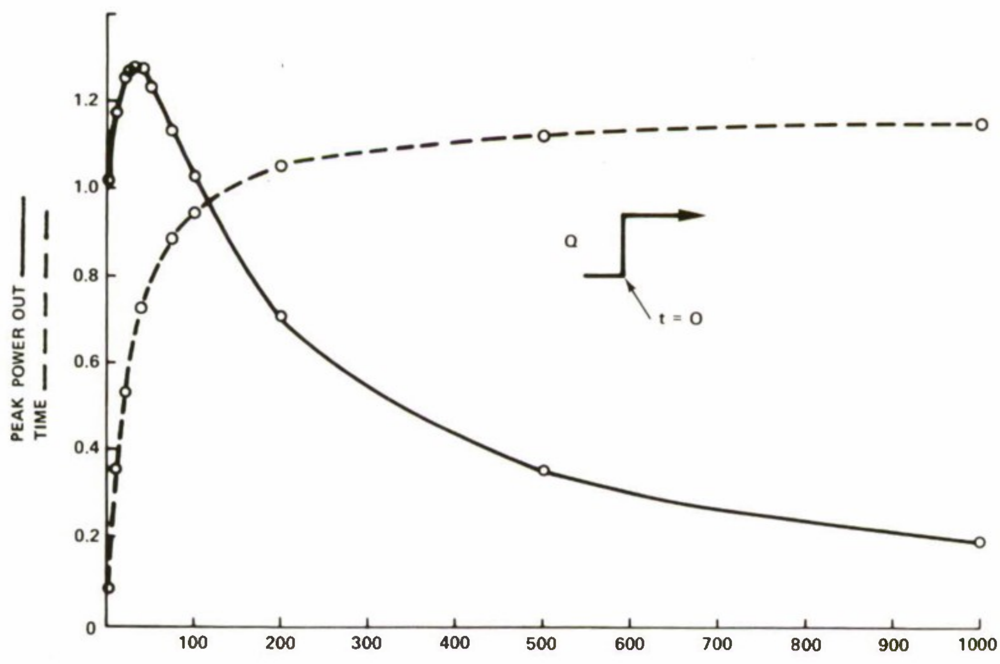

Figure 5. Peak output power (solid line) achieved at time (dashed line) as functions of Q in type A Q-switching.

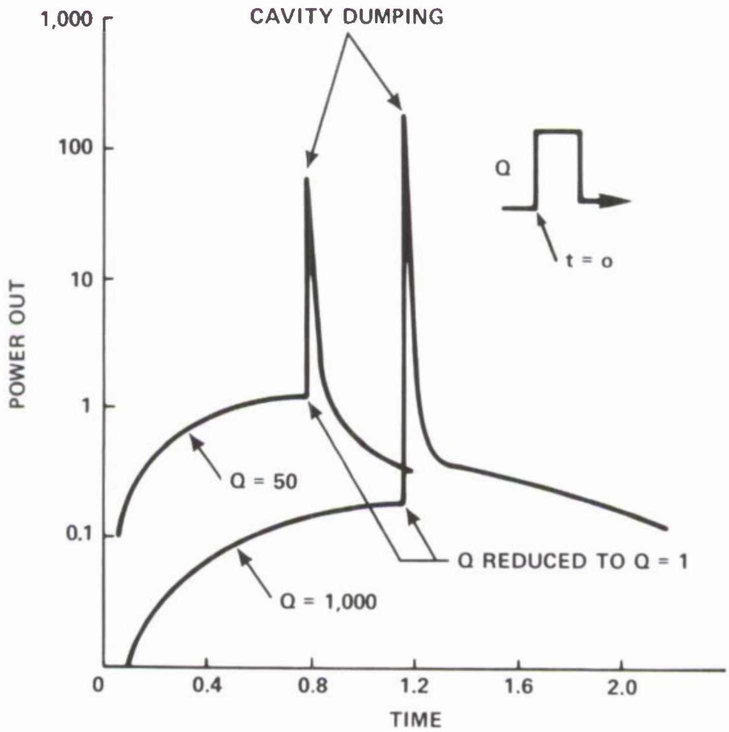

Figure 6. Power out versus time for two different Q in type B Q-switching.

Figure 6 shows the time variation of output power for Q-switching of type B. A large spike (cavity dumping) occurs when the Q is instantaneously lowered after the cavity laser power has reached maximum. The average output power keeps increasing with Q for this type of Q-switching, till it saturates at some finite Q when, presumably, all the energy that can be retrieved from the system has been retrieved. The maximum average output power and the time over which it is averaged are plotted as functions of Q in figure 7. The product of the two curves gives the total energy output in that time.

The values chosen for the system's parameters for these calculations are given in appendix A. It is *cautioned* that no attempt has been made in this preliminary report to assign physically meaningful numbers to the parameters. The numbers were chosen to display the properties of the model for convenient values of Q, time, and power.

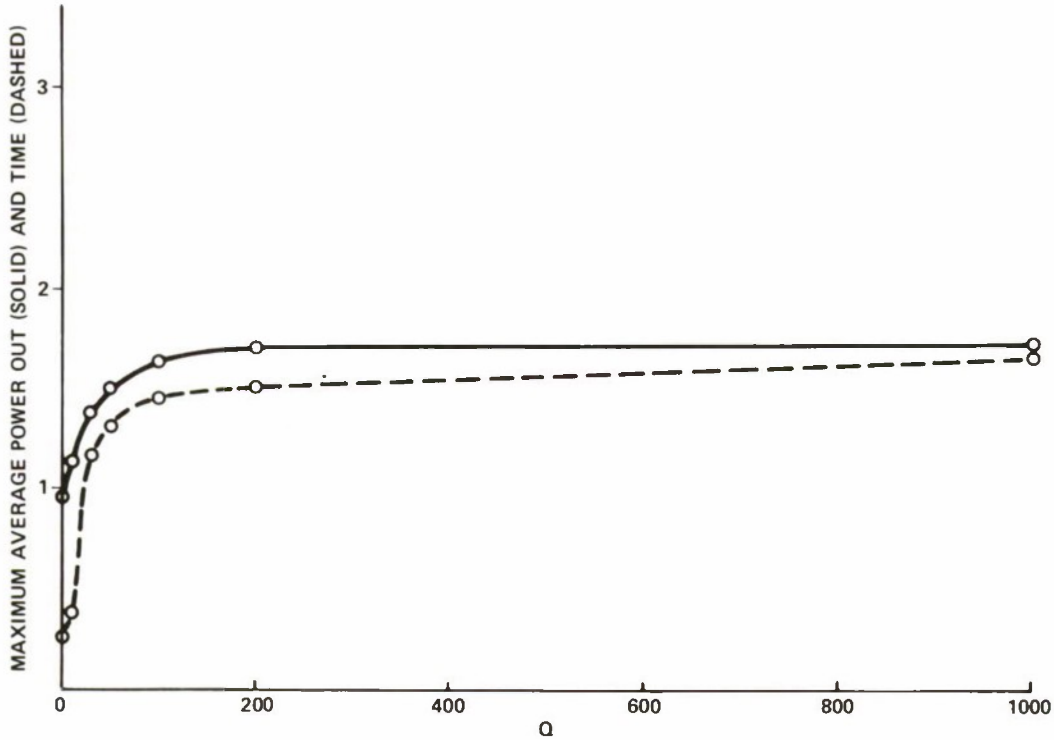

Figure 7. Maximum average power out and time over which it is delivered as functions of Q for type B Q-switching (energy out is product of two curves).

APPENDIX A.--PARAMETER VALUES

Values for the parameters used in these calculations are

$\nu_{21} = 1$	$\alpha_{31} = 3$
$kT = 1$	$\alpha_{32} = 3$
$\nu_{32} = 100$	$\alpha_{54} = 3$
$b_{32} = 100$	$s_{21} = 100d$
$\eta = 0.2$	$s_{31} = s_{32} = s_{54} = d$
$c_A = 0.1$	$d = \left[1 - (0.7)^6\right]^{-1} = 1.13333$
$c_B = 0.6$	$a_{21} = s_{21}\left[\exp(\nu_{21}/kT) - 1\right]^{-1}$
$x = 10$	
$M = 6$	

With these values, we obtained

$$s_{31} = s_{32} = s_{54} = 1.1334,$$

$$s_{21} = 113.334,$$

$$a_{21} = 65.9575,$$

$$Q_c = 37.9026 .$$

APPENDIX A

At zero pump power, the relative populations of the various levels are

$$f_3 = f_5 = 0 ,$$

$$f_4 = 1 ,$$

$$f_2 = 0.268941 ,$$

$$f_1 = 0.731059 .$$

There is substantial occupation of the terminal laser level (level 2), owing to the relatively small value of $\nu_{21}/kT = 1$ chosen above.

The values at $t = 0$ in the Q-switching modes that are established at $Q = 0$ and $P = P_o = 2.07445$ prior to $t = 0$ are

$$u_o = 0.277819 ,$$

$$v_o = 0.412911 ,$$

$$w_o = 0.444749 ,$$

$$z_o = 0 .$$

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